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KINEMATIC SHEAR VISCOSITY OF LIQUID ALKALINE METALS

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The origin of kinematic shear viscosity in liquid alkaline metals has been studied. It is shown that, since the depth of the well in the potential of pair interaction between ions is small in comparison with the energy of thermal motion of those ions, the mechanism of kinematic shear viscosity formation is not an activation one. The main mechanism consists in the momentum transfer from one layer to another and depends on the layer "roughness". In accordance with the generalized similarity principle, liquid alkaline metals are shown to belong to the same similarity class, and a similar character of changes in the isobars of the kinematic shear viscosity of liquid alkaline metals is observed only if this principle is applicable. A formula for the kinematic shear viscosity is proposed. The agreement of the results obtained with experimental data is quite satisfactory.

Keywords: kinematic shear viscosity, liquid alkaline metals.

1. Introduction

Researches of the dynamic viscosity in pure liquid metals and their alloys are a matter of substantial interest from the viewpoint of their application; in particular, as heat-carriers in nuclear power plants. A considerable number of experimental works (see, e.g., [1–6]) are devoted to this issue. As a rule, the dynamic viscosity η as a function of the temperature T is described with the help of Arrhenius law

$$\eta = \eta_1 \exp\left(\frac{E_a}{k_B T}\right), \quad (1)$$

where the parameter E_a is called the activation energy of viscous flow. But from the very beginning, the attention should be attracted to the magnitude of the activation energy of thermal motion. The ratios between the latter and the energy of thermal motion of atoms calculated at the melting point T_m are quoted in Table 1.

Formula (1) corresponds to a scenario with a crystal-like character of the thermal motion in liquid metals. In other words, an ion oscillates near a temporary equilibrium position during the time interval τ_0 ; afterward, within the time interval $\tau_1 \ll \tau_0$, it jumps into another temporary equilibrium position.

At the same time, the correct value of the activation energy of viscous flow should be determined from

Table 1. Dimensionless activation energy of viscous flow $\tilde{E}_a = E_a/(k_B T_m)$ and dimensionless depth of the potential well in the energy of interaction between two metal atoms $\tilde{U}_{\min} = U_{\min}/(k_B T_m)$

Atom	T_m , K	\tilde{E}_a	\tilde{U}_{\min}
Sn	504.9	1.50 [3]	~ 1.5
Bi	544.0	1.87 [3]	~ 1.5
Pb	600.4	1.56	~ 1.5
Li	452.9	1.19	~ 1.5
Na	370.8	2.44 [7]	1.35 [8]
K	336.7	1.82	1.49 [8]
Hg	311.9	1.56 [9]	

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isochores (see work [9]) rather than isobars, as was done in works [1–6] (see also Table 1). In agreement with work [9], the shear viscosity looks like

$$\eta = \eta_1 \exp\left(\frac{E_a(n)}{k_B T}\right),$$

where n is the density. The importance of this statement is illustrated by the behavior of the shear viscosity of argon, neon, and nitrogen (see Fig. 1). One can see that activation energies close to zero correspond to isochoric viscosity values. On isobars, those values are much larger.

Those qualitative speculations can be appended by a simple quantitative analysis. Let us proceed from the expression for the kinematic shear viscosity on isochores,

$$\nu = \nu_0 \exp(\varepsilon_a(n)/T),$$

where $\varepsilon_a(n) = E_a(n)/k_B$. At a displacement along any direction in the density-temperature plane (n, T), the shear viscosity changes according to the law

$$\frac{d\nu}{d\zeta} = \nu \left[\frac{1}{T} \frac{d\varepsilon_a(n)}{dn} - \frac{\varepsilon_a(n)}{T^2} \frac{dT}{d\zeta} \right], \quad (2)$$

where $d\zeta$ is an arbitrary linear combination of the density and temperature differentials. The effective energy of viscosity activation, which corresponds to this direction, is determined by the expression

$$\nu(\zeta) = \nu_0 \exp(\varepsilon_{\text{eff}}/T).$$

This yields

$$\frac{d\nu}{d\zeta} = -\frac{\varepsilon_{\text{eff}}}{T^2} \frac{dT}{d\zeta}. \quad (3)$$

Comparing Eqs. (2) and (3), we obtain

$$\varepsilon_{\text{eff}} = \varepsilon_a(n) - T \frac{d\varepsilon_a(n)}{dn} \left(\frac{dn}{d\zeta} / \frac{dT}{d\zeta} \right).$$

In particular, the isobaric value of activation energy equals

$$\varepsilon_{\text{eff}} = \varepsilon_a(n) - T \frac{d\varepsilon_a(n)}{dn} \left(\frac{dn}{dp} / \frac{dT}{dp} \right). \quad (4)$$

The data presented in Table 2 testify that the dimensionless values of activation energy on the isobars and the coexistence line considerably differ from

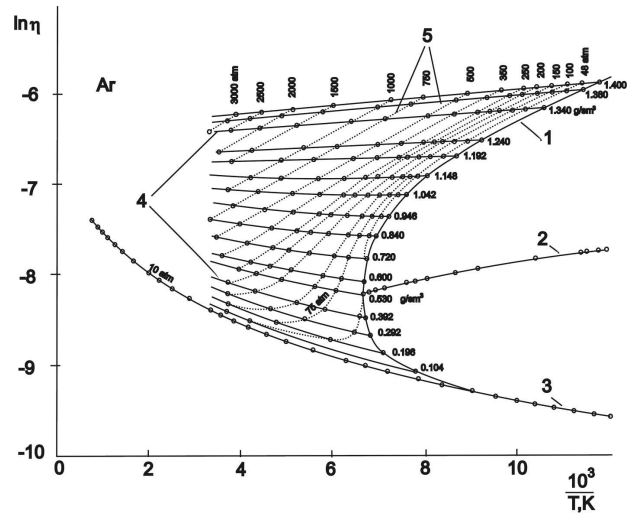


Fig. 1. Dependence of $\ln \eta$ on the reciprocal temperature, $1/T$, along the isochores and the coexistence curve for argon [10]: liquid phase (1), viscosity diameter (2), vapor-like phase (3), isochores (4), isobars (5). Points correspond to experimental values

Table 2. Dimensionless activation energy of argon on isochores and isobars and the coexistence curves

Ar				
$\rho = \text{const}$		$p = \text{const}$		Coexistence curve
$\rho, \text{ kg/m}^3$	$E_a/k_B T_c$	$P, \text{ MPa}$	$E_a/k_B T_c$	$E_a/k_B T_c$
600	-0.67	25	1.37	1.75
720	-5.55	50	1.25	
800	-0.46	75	1.17	
1000	-0.22	100	1.19	
1200	0.012			
1400	0.31			

the corresponding values on the isochores. Negative and small, in comparison with 1, values of the ratio $E_a/k_B T_c$ on the isochores point to a non-activation character of the viscous flow formation. At the same time, according to formula (4), $E_a/k_B T_c > 1$ on the isobars. In particular, substituting the corresponding values of the isochoric activation energy and the derivatives into Eq. (4), we obtain $\varepsilon_{\text{eff}} = 1.19$. Hence, the values of the “activation energy of viscous flow” that are measured experimentally on the isobars are

not true in the sense of the activation mechanism of shear viscosity formation in molecular liquids and liquid metals.

It should be noted that, as long ago as at the beginning of the 20th century, A. Bachynskyi proposed a “non-activation” formula for the shear viscosity [11]:

$$\eta = \frac{c}{v - v_0},$$

where v is the specific volume, and c a certain constant, which accounts for the intrinsic volume of a molecule, v_0 . However, this formula describes the temperature dependence of the shear viscosity rather well only in a restricted temperature interval.

The aim of this work was to consider the physical origin of the mechanism of shear viscosity formation in liquid alkaline metals. Let us start from the description of the kinematic shear viscosity rather than the dynamic one, because there is a necessity to exclude the influence of the density variation in the system with the temperature from consideration.

2. Principles of Viscosity of Atomic Liquids

Principles of the theory of kinematic viscosity in argon-like liquids were proposed in work [12]. The main result consists in that the viscosity of argon and all other low-molecular liquids, in which the averaged interaction potentials between molecules have an argon-like character, is described by the formula

$$\tilde{\nu}(v, t) = \frac{\zeta_0}{(\tilde{v} - \tilde{v}_0(t))^{1/3}}. \quad (5)$$

Here, the following notations are used: $\tilde{\nu} = \nu/\nu_R$ is the kinematic shear viscosity; ν_R the regularized value of kinematic shear viscosity, which is defined as $\nu_R = \nu(T_R)$; T_R the regularization temperature; $t = T/T_R$ the reduced temperature; $\tilde{v} = v/\nu_R$ the

regularized value of specific volume; and $v_R = v(t = 1) = v(T_R)$. The values of kinematic shear viscosity and specific volume are determined on the coexistence curve; they depend only on the temperature. The regularization temperature T_R divides the thermodynamic and fluctuation regions and has a meaning of the Ginzburg temperature. The numerical value of T_R is very close to the critical temperature. Let us also introduce the parameter λ ($\lambda < 1$) as the proportionality coefficient in the equality $v_R = \lambda v_c$, which makes it possible to exclude the influence of dimers, trimers, and higher-order clusters, which are formed near the critical point and substantially change the properties of the system [13, 14]. The other parameters are the excluded volume $v_0(t)$ and a certain constant ζ_0 .

Formula (5) successfully describes the kinematic shear viscosity of argon, krypton, and xenon, as well as liquid nitrogen, benzene, nitrobenzene, and many other low-molecular liquids (except for water and alcohols, in which rather strong hydrogen bonds play a substantial role). The most important qualitative feature of formula (5) consists in that the behavior of the shear viscosity mainly depends on the specific volume of a liquid (the viscosity value is almost constant along the isochores). Furthermore, formula (5) reflects the fact that the momentum transfer from one molecular layer to another that moves relatively to the former is associated with the layer “roughness” rather than the transfer of molecules between the layers.

Formula (5) can be rewritten in the form

$$\tilde{\nu}(\tilde{v}, t) \approx \frac{(1 - \tilde{v}_0)^{1/3}}{(\tilde{v} - \tilde{v}_0)^{1/3}}, \quad \zeta_0 = (1 - \tilde{v}_0)^{1/3}. \quad (6)$$

It takes into account that the left- and right-hand sides of formula (5) have to tend to 1 if the temperature $T \rightarrow T_R$. A comparison of the numerical values of key parameters is made in Table 3. One should pay attention that the values of the excluded volume v_0 and the corresponding values of v_{tr} are practically identical. In addition, the difference between the values of ζ_0 for various liquids does not go beyond the limits of experimental errors.

The approach to the viscosity problem, which was formulated in work [12], allows us to give a new formulation of the similarity theory, which considerably generalizes standard results [15, 16]. According to works [15, 16], similar are those states of the system, for which the values of the normalized temper-

Table 3. Parameters t_R , ν_R , v_0 , v_{tr} , \tilde{v}_0 , and ζ_0 for argon, nitrogen, and benzene

Parameter	Ar	N ₂	C ₆ H ₆
t_R	0.997	0.978	0.996
$\nu_R, 10^{-3} \text{ cm}^2/\text{s}$	0.519	0.61	1.18
$v_0, \text{ \AA}^3$	46.50	54.94	153.35
$v_{tr}, \text{ \AA}^3$	47.08	53.84	145.09
\tilde{v}_0	0.461	0.446	0.421
ζ_0	0.814	0.821	0.834

ature, $\bar{t} = \frac{T}{T_c}$, volume, $\bar{v} = \frac{v}{v_c}$, and pressure, $\bar{p} = \frac{P}{P_c}$, where the superscript c marks the corresponding value at the critical point, are identical. This formulation is quite correct for systems, in which the ratios $\frac{T_{tr}}{T_c}$ and $\frac{v_{tr}}{v_c}$ acquire identical values. The simplest example of such systems includes atomic liquids (neon, argon, krypton, xenon). All the values of relevant parameters are identical for those liquids and are quoted in Table 3. Furthermore, the dimensionless $\frac{T_{tr}}{T_c}$ - and $\frac{v_{tr}}{v_c}$ -intervals of existence for liquid states are also identical.

At the same time, for the majority of low-molecular systems, the temperature and volume intervals of existence for the liquid state substantially differ from each other. In this case, in accordance with work [17], the change to new generalized variables is a natural generalization of the standard similarity principle:

$$\frac{T}{T_c} \rightarrow \frac{T - T_{tr}}{T_c - T_{tr}}, \quad \frac{v}{v_c} \rightarrow \frac{v - v_{tr}}{v_c - v_{tr}}.$$

The character of the generalized similarity law for the kinematic shear viscosity was discussed in work [12]. It was shown that the kinematic shear viscosity should be described in the variables

$$\tilde{\nu} = \frac{\nu}{\nu_R}, \quad \tilde{v} = \frac{v - v_{tr}}{v_c - v_{tr}}.$$

This circumstance is illustrated in Fig. 2.

3. The Temperature Dependence of Viscosity in Liquid Alkaline Metals

While considering liquid metals, the same normalized variables as in the case of argon-like liquids will be used. Unfortunately, the determination accuracy for the critical parameters of alkaline metals is considerably lower (the errors can be up to 10%). Therefore, the regularized value of kinematic shear viscosity will be determined with the help of the interpolation into the critical region. This procedure is illustrated in Fig. 3. The values obtained for ν_R are quoted in Table 4.

Table 4. Regularized kinematic shear viscosity for liquid Li, Na, and K

Parameter	Li	Na	K
$\nu_R \times (10^4 \text{ cm}^2/\text{s})$	3.52	1.81	1.65

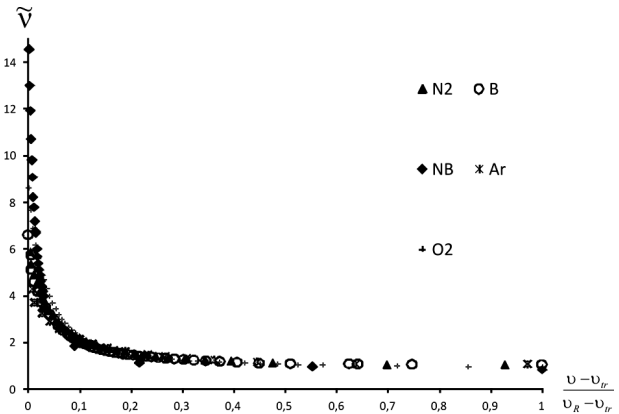


Fig. 2. Experimental data for the kinematic shear viscosity in argon, nitrogen, benzene, nitrobenzene, and oxygen (according to formula (7)) [12]

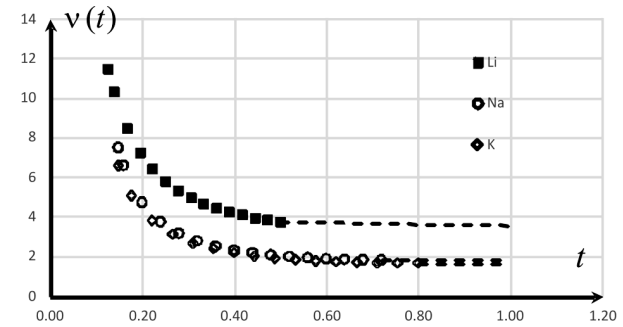


Fig. 3. Experimental data for the kinematic shear viscosity in liquid Li, Na, and K [18]

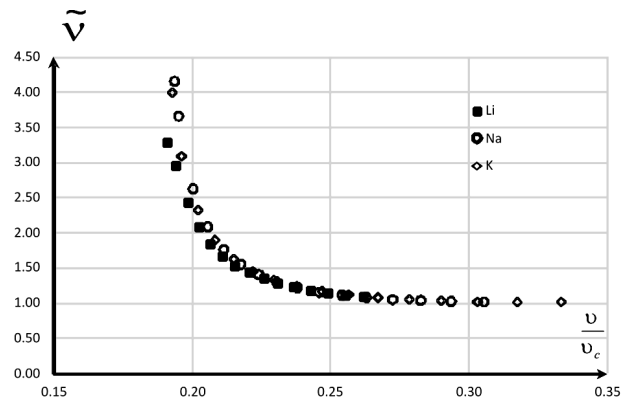


Fig. 4. Dependence of the normalized kinematic shear viscosity on v/v_c

The regularized value of specific volume was put equal to v_c . Figures 4 and 5 demonstrate the normalized kinematic shear viscosity of alkaline metals, $\tilde{\nu} = \frac{\nu}{\nu_R}$, in the standard, $\tilde{v} = \frac{v}{v_c}$, and generalized,

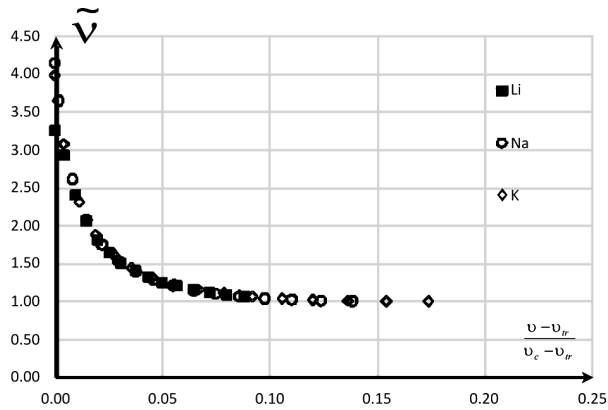


Fig. 5. Dependence of the normalized kinematic shear viscosity on $\frac{v - v_{tr}}{v_c - v_{tr}}$

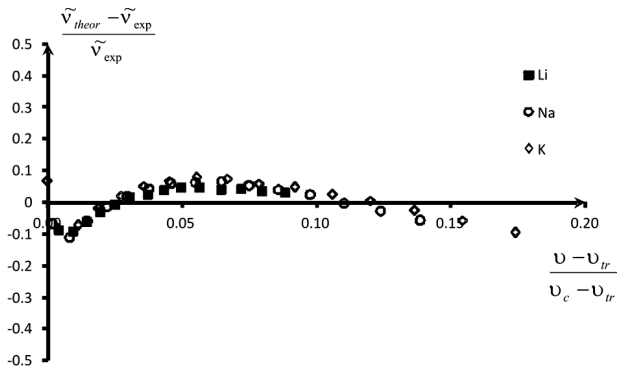


Fig. 6. Relative deviation $\frac{\tilde{v}_{theor} - \tilde{v}_{exp}}{\tilde{v}_{exp}}$ of the calculated values \tilde{v}_{theor} for the kinematic shear viscosity in alkaline metals by formula (8) from the corresponding experimental values \tilde{v}_{exp} [18]

$\tilde{v} = \frac{v - v_{tr}}{v_c - v_{tr}}$, variables. Comparing those figures, one can see that the best agreement of the data for various alkaline metals is observed (the points lie on a single curve) if the kinematic shear viscosity is considered as a function of $\tilde{v} = \frac{v - v_{tr}}{v_c - v_{tr}}$. This approach allows us to reveal a complete similarity of the kinematic shear viscosities for various liquid alkaline metals.

The difference between liquid alkaline metals and atomic liquids manifests itself through the values of dimensionless ratios $\frac{T_{tr}}{T_c}$ and $\frac{v_{tr}}{v_c}$ (see Table 5). From the tabulated data, it follows that the corresponding values of those parameters for Ar-like atomic liquids and liquid alkaline metals differ from one another by several times. At the same time, the character of the dependence of the kinematic shear viscosity in liquid alkaline metals on the specific volume turns out

identical. From this fact, it follows that atomic liquids and liquid alkaline metals should be classed to different similarity classes. This is a consequence of the fact that the pair interaction potentials in liquid metals substantially depend on the concentration [7, 15, 16].

The values determined by the least-square method for the quantities ζ_0 and \bar{v}_0 are quoted in Table 6. It follows that, within the experimental error, the corresponding values of those parameters, as well as of \bar{v}_{tr} , are identical for various alkaline metals. At the same time, the values of ζ_0 for atomic liquids and alkaline metals have almost a two-fold difference. Moreover, one of the key relations, $\zeta_0 = (1 - \tilde{v}_0)^{1/3}$, is not obeyed for alkaline metals, although the value of the right-hand side turns out close to $(1 - \tilde{v}_0)^{1/3}$. This fact together with the dimensionless ratios t_{tr} and \bar{v}_{tr} allows a conclusion to be drawn that, on the basis of the viscosity and the equations of state, atomic liquids and alkaline metals belong to different classes of generalized similarity. Note that the deviation of the normalized kinematic viscosity \tilde{v} in alkaline metals calculated by formula (6) from experimental values does not exceed 4–6%, except in a small vicinity of the melting point (Fig. 6).

In principle, the considerably nonmonotonic behavior of the quantity $\frac{\tilde{v}_{theor} - \tilde{v}_{exp}}{\tilde{v}_{exp}}$ near the melting point can have two origins: 1) nucleation effects [22, 23] and 2) higher demands on the determination accuracy for the specific and own volumes of ions. In the second case, those quantities have to be known to the fourth digit after the decimal point. Really, if the experimental value $\bar{v}_{tr} = 0.1912$ is used for Li, the calculation

Table 5. Ratios T_{tr}/T_c and v_{tr}/v_c

Parameter	Ar	Li	Na	K
T_{tr}/T_c	0.55	0.13	0.15	0.15
v_{tr}/v_c	0.34	0.19	0.19	0.19

Table 6. Parameters for liquid alkaline metals

Parameter	Li	Na	K
ζ_0	0.4363	0.4568	0.4712
\bar{v}_0	0.1895	0.1930	0.1919
\bar{v}_{tr}	0.1912	0.1940	0.1932

error for $\frac{\bar{n}u_{\text{theor}} - \bar{v}_{\text{exp}}}{\bar{v}_{\text{exp}}}$ amounts to about 12%. In the case $\bar{v}_{\text{tr}} = 0.1902$, the calculation error for $\frac{\bar{v}_{\text{theor}} - \bar{v}_{\text{exp}}}{\bar{v}_{\text{exp}}}$ amounts to about 1%. This problem does not arise outside the region near the triple point.

4. Conclusions

To summarize, the main attention in this work is focused on the physical origin of the kinematic shear viscosity in liquid alkaline metals. The latter differ from the majority of other types of metals in that the free electrons in them are formed only by valence electrons, and there are no unfilled internal shells. Liquid lead, tin, and aluminum are the simplest examples of metals with unfilled internal shells.

Liquid alkaline metals are characterized by a substantial dependence of the potential well depth of ion-to-ion interaction on the concentration [7, 19–21]. However, this circumstance does not affect the character of the momentum transfer between the molecular layers moving relatively to one another. In the work, it is shown that the momentum transfer from layer to layer is mainly driven by effects associated with the “roughness” of molecular layers.

The momentum transfer from one molecular layer to another, which consists in a direct transition of ions between the layers, has a meaning probably only in a vicinity of the critical point. Far from the latter, this ionic motion becomes complicated, because the free volume in the system, i.e. the difference between the specific volume per ion and the own ion’s volume, is too small.

In order to confirm this statement, we quote the specific volumes of particles near the triple point and the melting point at the atmospheric pressure, as well as the volumes of ions calculated by the formula $v_0 = \frac{\pi}{6}\sigma^3$, where σ is the ion diameter, in Table 7. The latter was determined as a root of the equation

$$U(r)|_{r=\sigma} = 0, \quad (7)$$

Table 7. Specific and own ionic volumes near the triple point

Parameter	Na	K
$v_m, \text{\AA}^3$	41.14	78.39
$v_0, \text{\AA}^3$	38.22	64.64

where $U(r)$ is the potential of ion-to-ion interaction [24]. As one can see, the free volume does not exceed 6–7% for Na and 20% for liquid K. This means that the distance between the hard ionic cores does not exceed 2–5% of σ , which evidently complicates the translational motion of ions through the system. In this work, it is shown that a similar change in the kinematic shear viscosity of various alkaline metals along their isobars is observed only in the framework of the approach where the generalized principle of similarity formulated in work [12] is applied.

The temperature dependence of the kinematic shear viscosity can manifest itself only through the temperature dependence of the normalized excluded volume. According to estimates made in works [25, 26], the ion diameter changes following the law $\sigma^{-3}(T) = a - bT$. It should be noted that the value of σ that is determined from Eq. (7) considerably exceeds the hard core diameter, which is independent of the temperature.

It should be emphasized that the value of the dimensionless parameter ζ_0 is a discriminant, which classifies atomic liquids, alkaline metals, and, probably, other metals into similarity classes. The quantitative agreement with experimental data is observed in the whole liquid state domain. The relative deviation of the calculated values from experimental ones does not exceed 4–6%, except in a small vicinity of the melting point.

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КІНЕМАТИЧНА ЗСУВНА
В'ЯЗКІСТЬ РІДКИХ ЛУЖНИХ МЕТАЛІВ

Резюме

Досліджено природу кінематичної зсувної в'язкості рідких лужних металів. Показано, що внаслідок малої глибини ями парної міжіонної взаємодії, порівняно з енергією теплового руху іонів, механізм формування кінематичної зсувної в'язкості не є активаційним. Основним механізмом є перенос імпульсу від одного шару до другого, що залежить від "шорсткості" молекулярних шарів. Згідно з узагальненим принципом подібності показано, що рідкі лужні метали належать до одного класу подібності і подібний характер зміни кінематичної зсувної в'язкості різних лужних металів на їх ізобарах спостерігається тільки в рамках застосування цього принципу. Запропоновано формулу розрахунку кінематичної зсувної в'язкості. Узгодження з експериментальними даними є цілком задовільним.